

Possible Verification of Tilted Anisotropic Dirac Cone in α -(BEDT-TTF)₂ I₃ Using Interlayer Magnetoresistance

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It is proposed that the presence of a tilted and anisotropic Dirac cone can be verified using the interlayer magnetoresistance in the layered Dirac fermion system, which is realized in quasi-two-dimensional organic compound α -(BEDT-TTF)₂ I₃. Theoretical formula for the interlayer magnetoresistance is derived using the analytic Landau level wave functions and assuming local tunneling of electrons. It is shown that the resistivity as a function of the azimuthal angle of the magnetic field takes the maximum in the direction of the tilt if anisotropy of the Fermi velocity of the Dirac cone is small. The procedure is described to determine the parameters of the tilt and anisotropy.

KEYWORDS: α -(BEDT-TTF)₂I₃, Dirac cone, magnetoresistance

There is a group of condensed matter systems in which low-lying properties of the conduction electrons are described by a relativistic Dirac equation with the velocity of light replaced by the Fermi velocity. Remarkable physical phenomena of such Dirac fermions are clearly demonstrated in graphene, a monolayer of graphite.^{1,2} In the observed integer quantum Hall effect the plateaus in the Hall conductivity are characterized by not integers, N , but $N + 1/2$. The origin of the shift of $1/2$ is the presence of the zero energy Landau level for the Dirac fermions.

In contrast to graphene, which is a purely two-dimensional system, the organic conductor α -(BEDT-TTF)₂ I₃³ is the first bulk material where the massless Dirac fermion-like spectrum is realized. Experimentally it was reported that this quasi-two-dimensional compound was a narrow gap system under high pressure.⁴⁻⁷ Using the tight-binding model with the transfer integrals obtained by X-ray diffraction experiment,⁸ Kobayashi *et al.* suggested that the system is zero-gap and the energy dispersion is linear around the zero-gap point.^{9,10} They showed that the electronic band structure is described by a tilted and anisotropic Dirac cone. The first principle calculations supported this Dirac cone structure.^{11,12} Although observed negative interlayer magnetoresistance¹³ is an evidence of the presence of a Dirac cone, as theoretically explained by Osada¹⁴ in terms of the zero energy Landau level wave function, there is no direct experimental verification that the cone is tilted and anisotropic.

In this Letter, we propose that the Dirac cone structure is verified by analyzing interlayer magnetoresistance through its dependence on the applied magnetic field direction. Using the analytic form of the zero-energy Landau level wave function with tilt and anisotropy of the Dirac cone,¹⁵ we derive a formula

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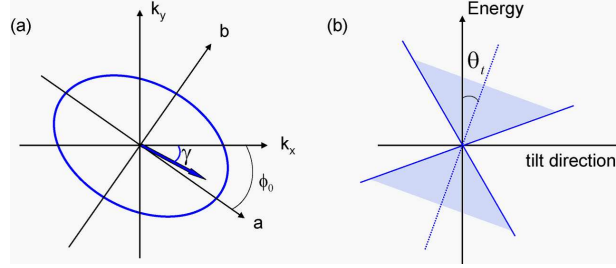


Fig. 1. (a) The definition of the crystal axes denoted by a and b and the wave vector axes k_x and k_y . The angle between the a -axis and the k_x -axis is represented by ϕ_0 . The arrow indicates the tilt direction. The angle between the k_x -axis and the tilt direction is defined as γ . By tilting the cone the intersection becomes an ellipse whose long axis is in the tilt direction as schematically shown in the figure. (b) The definition of the tilt angle, θ_t . The horizontal axis is the tilt direction in the k_x - k_y plane. The vertical axis is the energy axis. The declined solid lines represent the dispersion of the Dirac cone along the tilt direction. The dotted line represents the center line of the two dispersions. The angle between the energy axis and the center line is θ_t .

for the interlayer magnetoresistance. Our formula is unique for the analysis of the tilted and anisotropic Dirac cone in α -(BEDT-TTF) $_2$ I $_3$. It should be noted that in α -(BEDT-TTF) $_2$ I $_3$ Shubnikov-de Haas oscillations, which is a standard experiment to study an electronic structure, have never been observed and angle resolved photoemission spectroscopy, which directly confirmed the linear dispersion in graphene,¹⁶ is not applicable to organic compounds.

Before beginning the analysis of the system, we introduce parameters defining the tilt and anisotropy of the Dirac cone. We represent the crystal axes in the plane as a and b . We assume that the intersections of an anisotropic Dirac cone are elliptic. The principal axes are not necessarily parallel to the crystal axes. So the k_x - and k_y -axes in the wave vector space are taken so that those axes are parallel to the principal axes. The angle between the k_x -axis and the a -axis is denoted by ϕ_0 as shown in Fig.1(a). As for the tilt direction, we introduce γ to denote the angle between the k_x -axis and the tilt direction. The parameter θ_t describes the tilt angle as defined in Fig.1(b). Thus, the parameters are γ , θ_t , and the Fermi velocity anisotropy (denoted by α below). Within our formulation, ϕ_0 is taken as a given parameter.

Now we start with the Hamiltonian describing a single layer of an anisotropic and tilted Dirac cone system following Ref.17,

$$H = \sum_{k_x, k_y} H(k_x, k_y), \quad (1)$$

where

$$H(k_x, k_y) = \hbar \begin{pmatrix} v_0^x k_x + v_0^y k_y & v_x k_x - i v_y k_y \\ v_x k_x + i v_y k_y & v_0^x k_x + v_0^y k_y \end{pmatrix}. \quad (2)$$

This is a sufficiently general form. Anisotropy in the Fermi velocity is parameterized by $\alpha = \sqrt{v_x/v_y}$. In the absence of the magnetic field the energy dispersion is given by $\epsilon_k = \hbar \left(v_0^x k_x + v_0^y k_y \pm \sqrt{v_x^2 k_x^2 + v_y^2 k_y^2} \right)$.

The angle γ defined in Fig.1(a) is given by

$$\gamma = \cot^{-1} \frac{v_0^x v_y}{v_0^y v_x}. \quad (3)$$

To compute the interlayer magnetoresistance, we need the Landau level wave functions. We represent the magnetic field as $(B_x, B_y, B_z) = B(\cos \theta \cos \phi, \cos \theta \sin \phi, \sin \theta)$. Here ϕ is the angle in the plane with respect to the positive k_x -axis and θ is the angle between the magnetic field direction and the plane. We choose the gauge so that the vector potential is given by $A_x = B_y z + A_x^{(z)}$, $A_y = -B_x z + A_y^{(z)}$, and $A_z = 0$ where $B_z = \partial_x A_y^{(z)} - \partial_y A_x^{(z)}$. The presence of the inplane magnetic field is taken into account by a gauge transformation because they depend only on z . The zero-energy Landau level wave function is given in Ref.15. Here we derive it in a different way which can be applicable to compute other Landau level wave functions. The derivation procedure consists of three steps. After the Pierls substitution $(k_x, k_y) \rightarrow (\kappa_x, \kappa_y) = (k_x + eA_x^{(z)}/c\hbar, k_y + eA_y^{(z)}/c\hbar)$, first we rescale k_y so that the Fermi velocity in this direction is v_x . At this transformation, B_z is multiplied by v_y/v_x . Secondly we rotate the system by the angle γ in the plane. The transformed Hamiltonian is,

$$H(\kappa_x, \kappa_y) = \hbar v_x U^\dagger (-\eta \kappa_x \sigma_0 + \kappa_x \sigma_1 + \kappa_y \sigma_2) U, \quad (4)$$

where κ_x and κ_y are redefined as the rotated variables, σ_j ($j = 1, 2$) are the Pauli matrices and

$$\eta = \sqrt{\left(\frac{v_0^x}{v_x}\right)^2 + \left(\frac{v_0^y}{v_y}\right)^2}, \quad (5)$$

$$U = \begin{pmatrix} \exp(\frac{i}{2}\gamma) & 0 \\ 0 & \exp(-\frac{i}{2}\gamma) \end{pmatrix}. \quad (6)$$

Now the direction of tilt is in the κ_x -axis. The angle of tilt is defined by $\theta_t = \frac{\theta_+ - \theta_-}{2}$, where $\tan \theta_\pm = 1 \pm \eta$. In the Schrödinger equation with the Hamiltonian UHU^\dagger , we subtract the term associated with the tilt from the both side of the equation. Applying the operator of the Dirac cone and after some algebra we obtain the operator form of an anisotropic harmonic oscillator. Diagonalizing the operator, we find the energies of the Landau levels,

$$\epsilon_n = \text{sgn}(n) (\hbar v_x / \ell_z) \sqrt{2\lambda^3 |n|} \quad (7)$$

with $n = 0, \pm 1, \pm 2, \dots$, the magnetic length $\ell_z = \sqrt{c\hbar/eB_z}$ and

$$\lambda = \sqrt{1 - \eta^2} = \sqrt{1 - \left(\frac{v_0^x}{v_x}\right)^2 - \left(\frac{v_0^y}{v_y}\right)^2}. \quad (8)$$

The Landau level wave functions are obtained by taking the Landau gauge for $(A_x^{(z)}, A_y^{(z)})$,

$$\begin{aligned} \phi_n(\zeta_k^{(n)}) &= \frac{1}{\sqrt{4(1+\lambda)}} \left[(1 - \delta_{n,0}) \begin{pmatrix} 1 + \lambda \\ \eta \end{pmatrix} f_{|n|-1}(\zeta_k^{(n)}) \right. \\ &\quad \left. + \text{sgn}(n) \begin{pmatrix} \eta \\ 1 + \lambda \end{pmatrix} f_{|n|}(\zeta_k^{(n)}) \right], \end{aligned} \quad (9)$$

with $\text{sgn}(0) = 1$. Here k is the wave vector for the plane wave component, where the plane wave part is implicitly included, and

$$\zeta_k^{(n)} = \sqrt{\lambda} \left[\frac{\alpha}{\ell_z} (x \sin \gamma + y \cos \gamma) + \frac{\ell_z}{\alpha} k \right] - \eta \sqrt{2|n|} \text{sgn}(n), \quad (10)$$

$$f_n(\zeta) = \frac{(-1)^n}{2^{n/2} \pi^{1/4} \sqrt{n!}} H_n(\zeta) \exp\left(-\frac{1}{2}\zeta^2\right), \quad (11)$$

with $H_n(\zeta)$ the Hermite functions.

The interlayer conductivity is calculated using the Kubo formula. As for the impurity scattering, we assume that the scattering leads to Lorentzian-shape density of states with half value width of a constant Γ . Although a self-consistent Born approximation shows that Γ is magnetic field dependent,¹⁸ here we ignore field dependence of Γ for simplicity. We focus on the contribution from the zero energy Landau level at zero temperature. We shall comment on the effect of the other Landau levels later. The matrix elements of the current operator is calculated similarly to the non-tilted case¹⁴ assuming local tunneling of electrons between two neighboring layers.¹⁹ The interlayer magnetoresistance is given by

$$\rho_{zz}^{(0)} = \frac{A}{B_0 + B \sin \theta \exp \left[-\frac{1}{2} \left(\frac{a_c}{\ell_z} \right)^2 \frac{\cos^2 \theta}{\sin^2 \theta} I(\phi, \alpha, \gamma, \lambda) \right]}, \quad (12)$$

where

$$\begin{aligned} I(\phi, \alpha, \gamma, \lambda) &= \lambda \left(\alpha \sin \phi \cos \gamma - \frac{1}{\alpha} \cos \phi \sin \gamma \right)^2 \\ &+ \frac{1}{\lambda} \left(\alpha \sin \phi \sin \gamma + \frac{1}{\alpha} \cos \phi \cos \gamma \right)^2. \end{aligned} \quad (13)$$

Here B_0 is a parameter of the theory and $A = \frac{\hbar}{e^2} \left(\frac{2\pi^2 \Gamma^2}{N_c t_c^2} \right) \left(\frac{c\hbar}{ea_c^2} \right)$ is taken to be a constant with N_c the number of layers, t_c the hopping parameter between neighboring layers, and a_c the lattice constant perpendicular to the plane. Note that the function $I(\phi, \alpha, \gamma, \lambda)$ has the period π with respect to ϕ as confirmed analytically. Note also that there is another Dirac cone with $\eta \rightarrow -\eta$ by symmetry.¹⁷ But that Dirac cone has the same contribution because the expression is invariant under this sign change of η .

Let us move on to the parameter dependence of the interlayer magnetoresistance. Figure 2 shows the angular ϕ dependence of $\delta\rho_{zz}^{(0)} = -[\rho_{zz}^{(0)} - \rho_0]/\rho_0$ for various values of λ with $\alpha = 1$ ($v_x = v_y$). Here ρ_0 is the interlayer resistance in the absence of the magnetic field. On the other hand, Fig. 3 shows the angular ϕ dependence for various values of α in the absence of the tilt. Anisotropy of the Fermi velocity also leads to an angular dependence of the interlayer magnetoresistance. As for the θ dependence, we found that the ratio of the maximum to the minimum in Figs. 2 and 3 decreases with increasing θ .

The fact that anisotropy in eq.(12) in the $x - y$ plane arises from the tilt is understood as follows. In the presence of tilt, the intersection of the Dirac cone is deformed. In case of $\eta = 0$ and $v_x = v_y$, the intersection is circle. But if $\eta \neq 0$, the intersection becomes an ellipse with the origin at $(k_x^{(n)}, k_y^{(n)}) = (\eta(\epsilon_n/\hbar v_x)/(1 - \eta^2), 0)$ and the ratio of the principal axes being λ . The form of the Landau level wave functions is deformed according to this change of the intersection. We find that the Landau level wave

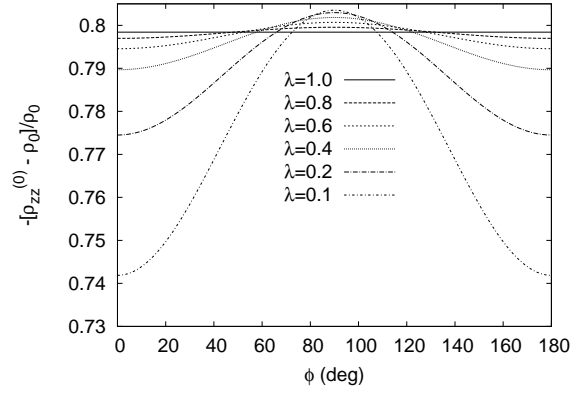


Fig. 2. Dependence of the interlayer magnetoresistance on ϕ for various tilt parameters λ at $B = 6\text{T}$, $B_0 = 0.5\text{T}$ and $\theta = 20$ degrees with $\alpha = 1 (v_x = v_y)$.

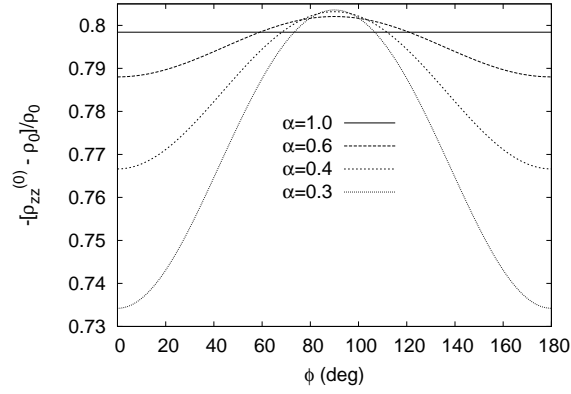


Fig. 3. Dependence of the interlayer magnetoresistance on ϕ in the absence of the tilt ($\lambda = 1$) for various α values. Other parameters are the same as in Fig.2.

function is more localized in the tilt direction than the perpendicular direction to the tilt direction. Since the matrix element of the interlayer current operator has a Fourier transformation like form with respect to the in-plane magnetic field, the current operator matrix element takes the minimum if the in-plane magnetic field is in the tilt direction. Therefore, the interlayer resistivity takes the maximum if the in-plane magnetic field is in the tilt direction.

Kobayashi *et al.* estimated the parameters of the anisotropic tilted Dirac cone at the uniaxial pressure $P_a = 4.5\text{kbar}$ along the a -axis.¹⁷ Using those values, we find $\gamma = -31.6$ degrees, $\eta = 0.92$, $\lambda = 0.40$, $\alpha = 1.18$, and $\phi_0 = 32.6$ degrees. Figure 4 shows the interlayer magnetoresistance for this parameter set. From the comparisons with the no tilt case ($\lambda = 1$, $\alpha = 1.18$) and the isotropic Fermi velocity case ($\lambda = 0.40$, $\alpha = 1.0$) with using the same other parameter values, we see that the ϕ -dependent interlayer magnetoresistance comes from the tilt of the Dirac cone.

Experimentally a rough estimation of the tilt angle, γ , is obtained if we assume $\alpha \simeq 1$. In this case

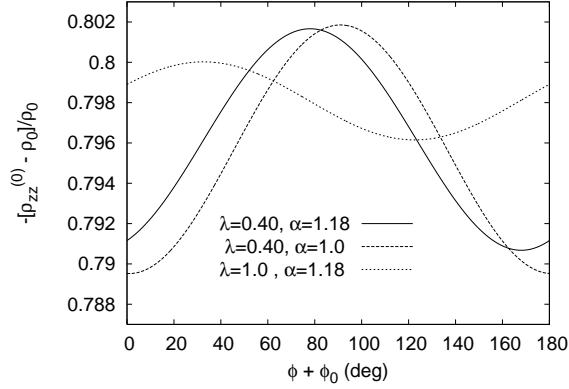


Fig. 4. Dependence of the interlayer magnetoresistance on ϕ using the parameters estimated by Kobayashi *et al.*¹⁷ at $P_a = 4.5$ kbar. Other parameters are set as $B = 6$ T, $B_0 = 0.5$ T, and $\theta = 20$ degrees. The results for no tilting case and isotropic Fermi velocity case are shown as well.

anisotropy of the interlayer magnetoresistance in the plane is mainly associated with the tilt. For $\alpha = 1$, the function I takes a simple form,

$$I(\phi, \alpha = 1, \gamma, \lambda) = \frac{1}{2} \left(\frac{1}{\lambda} + \lambda \right) + \frac{1}{2} \left(\frac{1}{\lambda} - \lambda \right) \cos 2(\phi - \gamma). \quad (14)$$

From this expression one can see that the resistivity takes the maximum in the direction of the tilt. An approximate value of γ is found from the angle of that direction. The other parameters λ and α can be estimated as follows. We first extract the function $I(\phi, \alpha, \gamma, \lambda)$ part from the experimental data using the parameters A and B_0 determined from θ dependence of $\rho_{zz}^{(0)}$. Taking the approximate values of γ and λ , which is obtained by using eq.(14), and $\alpha = 1$ as initial values, more precise values are determined by the least squares method.

Now we comment on the conditions for the application of the formula (12). The formula is derived by using the zero-energy Landau level wave function. To justify this approximation, the magnetic field B_z should be large enough. The energy gap to the first excited Landau level is $\epsilon_1 \simeq 40\sqrt{B_z}$ K where B_z is measured in units of tesla if we assume the average Fermi velocity $\simeq 10^7$ cm/s.¹³ Therefore, if the temperature is sufficiently lower than ϵ_1 then the effect of the other Landau levels is safely neglected. As for the angle θ , the condition is $40\sqrt{B \sin \theta} > k_B T$. The formula including the effect of the other Landau levels is necessary for small B_z values. It is straightforward to extend the formula to this case. But the formula is complicated and such a formula is necessary only when one tries to see the crossover from the quantum limit to the semi-classical regime. However, this is beyond the scope of this paper. Another effect to be concerned is the Zeeman splitting.¹⁴ However, the Zeeman splitting leads to a constant shift of the energy even though the shift depends on the spin. At fixed total magnetic field B the shift is unimportant for the determination of the parameters of the Dirac cone because it just leads to a modification of A in eq.(12).

To conclude, we have derived the formula for the interlayer magnetoresistance in the presence of the

tilt and the Fermi velocity anisotropy of the Dirac cone. The direction of the tilt is determined from the azimuthal angle dependence of the interlayer magnetoresistance. If the interlayer resistivity takes the maximum in some direction, then the Dirac cone is tilted in that direction. Physically the resistivity takes the maximum in the tilt direction because the interlayer current operator matrix element takes the minimum. The derived formula can be used to extract pressure dependence of the parameters of the tilted and anisotropic Dirac cone. It would be interesting to see the difference between the parameters determined by applying the formula to analyze the experimental data and the band calculation result. We expect that discrepancy between them should be large under low pressures because the Dirac fermions become unstable due to the electron correlation leading to the charge ordering^{20–23} and/or superconductivity.^{9,24}

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